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Palasantzas, George

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# Static and dynamic aspects of the rms local slope of growing random surfaces

George Palasantzas

*Department of Applied Physics, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands*

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In this work, we investigated static and dynamic aspects of the rms local surface slope “ $\rho$ ” for self-affine random surfaces. The rms local slope is expressed as a function of the rms roughness amplitude  $\sigma$ , the in-plane correlation length  $\xi$ , and the roughness exponent  $H$  ( $0 < H < 1$ ), as well as is shown to scale as  $\rho \sim \sigma \xi^{-H}$ . Application to room temperature heteroepitaxial silver films shows the rms local slope to be closely time invariant in the thickness range  $10 < h < 1000$  nm with an asymptotic value  $\rho \approx 0.7$ . However, discrepancies in deposition details could alter the mode of film growth leading to a power law growth of the local slope as a function of the film thickness  $h$ ;  $\rho \propto h^c$  ( $c > 0$ ). [S1063-651X(97)04007-5]

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The kinetic roughening of growing surfaces has been a topic of intensive research for the past ten years because of fundamental and technological importance [1,2]. The main growth factors of thin films under nonequilibrium conditions are deposition, desorption, and surface diffusion. A balance among them leads, in many cases, to the self-affine scaling hypothesis [3,4]. In terms of this hypothesis, the rms surface width grows with time  $t$  and length  $L$  as  $\sigma(L, t) = L^H F(t/L^{1/z})$  with  $\sigma(L) \propto L^H$ , if  $t/L^{1/z} \rightarrow +\infty$ , and  $\sigma(t) \propto t^\beta$ , if  $t/L^{1/z} \rightarrow 0$  [3]. The exponents  $H$ ,  $\beta$ , and  $z$  are, respectively, the roughness exponent [5], the growth exponent, and the dynamic exponent which describes the growth of the in-plane correlation length  $\xi$ ;  $\xi \propto t^{1/z}$ . Moreover, the scaling exponents obey the consistency relation  $z = H/\beta$  [3].

For surface diffusion driven growth where the desorption is negligible, nonlinear growth models predicted the exponents  $(H, \beta) = (2/3, 1/5)$  [6], while their linear versions yielded  $(H, \beta) = (1, 1/4)$  [7]. Monte Carlo simulations designed to describe the detailed microscopic processes of these equations [8] showed that the nonlinear models describe an intermediate range surface diffusion, while the linear ones describe a local surface diffusion. In the latter case, a groove instability develops with the rms local slope increasing with time as  $\rho \propto \ln^{1/2}(t)$  [9]. Such an anomalous scaling behavior was observed in a low temperature homoepitaxial growth on Si(111) [10], and in heteroepitaxial growth of Pt on glass [11], where the measured roughness exponent was found to be  $H \approx 0.9$ . By contrast, for nonlinear surface diffusion models [6] the rms local slope remains time invariant in the self-affine scaling regime [12].

In former heteroepitaxial studies where the growth process was surface diffusion driven (Ag/quartz) [13], the measured exponent  $H$  was found distinctly lower than 0.9, and larger than that predicted by the nonlinear diffusion models ( $> 2/3$ ) [6]. Moreover, the consistency relation  $z \approx H/\beta$  was experimentally confirmed, and it was shown that details of the deposition processes can have a great impact on the extent, spatial correlations can develop which lead to the discrepancy  $z \neq H/\beta$ . Nevertheless, the thickness dependence of the rms local surface slope remained unexplored, and will be the topic of the present work. Indeed, the temporal evolution of the rms local slope can be a unique feature to distinguish linear diffusion processes from the nonlinear ones. Further-

more, emphasis will be given to analytic calculations of the rms local surface slope as a function of the roughness parameters  $\sigma$ ,  $\xi$ , and  $H$  in terms of simple phenomenological correlation models which, however, can capture the correct self-affine asymptotic behavior and compare it with real data reasonably well [13,14].

The rms local surface slope is given by  $\rho = [\langle (\nabla z)^2 \rangle]^{1/2}$ , with  $z(r)$  the surface height profile [ $\langle z(r) \rangle = 0$ ]. The latter is considered a random (single valued) function of the in-plane position vector  $r = (x, y)$ . If we define the Fourier transform of  $z(r)$  by  $z(r) = \int z(q) e^{-iq \cdot r} d^2q$ , we obtain, after ensemble average over possible roughness realizations,

$$\rho = \left[ - \int \int q \cdot q' \langle z(q) z(q') \rangle e^{-i(q+q') \cdot r} d^2q d^2q' \right]^{1/2}. \quad (1)$$

For statistically stationary surfaces up to second order (translation invariance), we have  $\langle z(q) z(q') \rangle = [(2\pi)^4/A] \delta^2(q + q') \langle |z(q)|^2 \rangle$ . Upon substitution in Eq. (1) we obtain the rms local slope over an area of dimensions  $L \times L$

$$\rho(L) = \left[ \frac{(2\pi)^5}{A} \int_{k_L < q < Q_c} q^3 \langle |z(q)|^2 \rangle dq \right]^{1/2}, \quad (2)$$

where  $A$  is the macroscopic average flat surface,  $k_L = 2\pi/L$ , and  $Q_c = \pi/a_0$  with  $a_0$  the atomic spacing. For  $L \rightarrow +\infty$  ( $L \gg \xi$ ), Eq. (2) yields  $\rho(L) \approx \rho$ , with  $\rho$  rms local slope over an area with macroscopic dimensions.

For self-affine fractal surfaces, the roughness spectrum  $\langle |z(q)|^2 \rangle$  has the asymptotic scaling behavior [3]

$$\langle |z(q)|^2 \rangle = \begin{cases} \propto q^{-2-2H} & q\xi \gg 1 \\ \text{const} & q\xi \ll 1. \end{cases} \quad (3)$$

The asymptotic limits of  $\langle |z(q)|^2 \rangle$  in Eq. (3) are satisfied by the simple Lorentzian model  $\langle |z(q)|^2 \rangle_{sf} = [A/(2\pi)^5] \sigma^2 \xi^2 (1 + a q^2 \xi^2)^{-1-H}$  [14]. Indeed for  $q\xi \gg 1$ , we have  $\langle |z(q)|^2 \rangle_{sf} \propto q^{-2-2H}$ , while for  $q\xi \ll 1$ ,  $\langle |z(q)|^2 \rangle_{sf} \propto \sigma^2 \xi^2$ . The latter as a function of  $\xi$  reads of the form  $\langle |z(q)|^2 \rangle_{sf} \propto \xi^{2+2H}$ , since for growing self-affine surfaces  $\sigma \propto \xi^H$  [3]. The parameter “ $a$ ” is given by the relations  $a = (1/2H)[1 - (1 + a Q_c^2 \xi^2)^{-H}]$ , if  $0 < H < 1$ , and  $a$

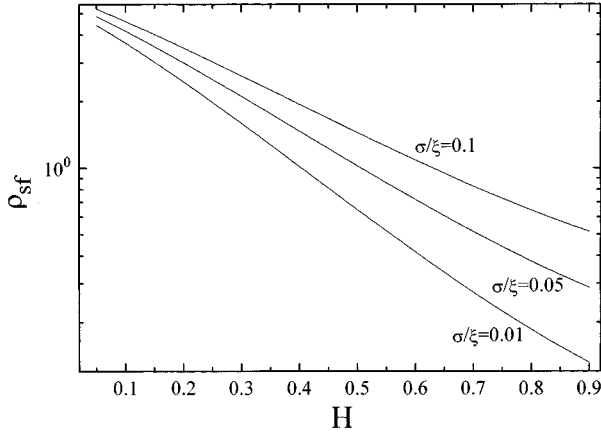


FIG. 1. Schematics of the rms local surface slope  $\rho_{sf}$  vs  $H$  (for  $L \rightarrow +\infty$ ) for  $a_0 = 0.3$  nm,  $\sigma = 2.0$  nm, and  $\xi = 20, 40, 80$  nm.

$= (1/2) \ln(1 + aQ_c^2 \xi^2)$ , if  $H = 0$  (logarithmic roughness) [14]. Besides the simplicity of  $\langle |z(q)|^2 \rangle_{sf}$ , its Fourier transform yields the analytically solvable correlation function  $C(r) = 2\sigma^2 - 2[\sigma^2/a\Gamma(1+H)](r/2a^{1/2})^H K_H(r/2a^{1/2}\xi)$  [14]. Finally, we point out that the roughness exponent  $H$  is a measure of the degree of surface irregularity [15], and is related with a local fractal dimension  $D = 3 - H$  [3].

Calculation of the rms local surface slope from Eq. (2) and the known expression of  $\langle |z(q)|^2 \rangle_{sf}$  yields

$$\rho_{sf}(L) = \frac{\sigma}{2^{1/2}a\xi} \left\{ \frac{1}{1-H} [X_c^{1-H} - X_L^{1-H}] + \frac{1}{H} [X_c^{-H} - X_L^{-H}] \right\}^{1/2}, \quad (4)$$

$$X_c = 1 + aQ_c^2 \xi^2 \quad \text{and} \quad X_L = 1 + ak_L^2 \xi^2.$$

For  $\xi \gg a_0$  and  $0 < H < 1$ , since  $Q_c \xi \gg 1$ , Eq. (4) for  $L \gg \xi$  yields the asymptotic behavior  $\rho_{sf} \approx B(H)(\sigma\xi^{-H})$ , with  $B(H) = [Q_c^{1-H}/a^H(2-2H)^{1/2}]$  which shows that the local slope scales primarily as  $\sim \sigma\xi^{-H}$ .

Figure 1 depicts the dependence of the local slope [Eq. (4) for  $L \rightarrow +\infty$ ] on the roughness exponent  $H$  for various values of the long-wavelength ratio  $\sigma/\xi$ . The effect of  $H$  on the local slope is dominant in comparison with that of  $\sigma/\xi$ . In fact, an increase of  $H$  from 0 to 1 leads to a decrement of the local slope even by two orders of magnitude, while an increment of  $\sigma/\xi$  by an order of magnitude cause only a moderate increment of the local slope which is more pronounced for large roughness exponents  $H(>0.5)$ . Finally, Fig. 2 displays the rather weak dependence of the local surface slope on the in-plane length scale  $l$  in terms of Eq. (4).

During the growth of self-affine surfaces,  $\sigma$  and  $\xi$  evolve as a function of film thickness (for constant deposition rate;  $t \sim h$ ) [3] as  $\sigma = A_1 h^\beta$ , and  $\xi = A_2 h^{1/2}$ , which upon substitution into Eq. (4) for  $L \gg \xi$ , we obtain  $\rho_{sf} \approx [B(H)A_1/A_2^H]h^{(\beta z - H)/H}$ . Thus, the local slope is time invariant  $\partial \rho_{sf} / \partial h = 0$  for  $H = \text{const}$ , if and only if  $\beta z = H$ . The application of kinetic growth theories to heteroepitaxial systems was concluded earlier on growth studies of CuCl on CaF<sub>2</sub>(111) [16], and the relation  $\beta z = H$  was confirmed ex-

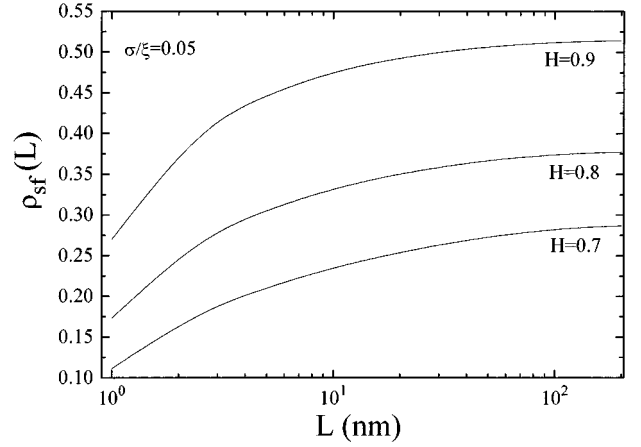


FIG. 2. Schematics of the rms local surface slope  $\rho_{sf}(L)$  vs  $L$  for  $a_0 = 0.3$  nm,  $\sigma = 2.0$  nm,  $\xi = 40$  nm, and three consecutive values of the roughness exponent  $H$  ( $=0.7, 0.8, 0.9$ ).

perimentally for room temperature grown Ag/quartz films [13,17]. Using the experimentally determined relations  $\sigma = (0.956)h^{0.29}$  and  $\xi = (5.31)h^{0.40}$  ( $h$  in nm) [13,17] and Eq. (4) for  $L \rightarrow +\infty$  with  $H = 0.82$ , we calculated in Fig. 3 the local slope as a function of film thickness. The latter decreases with film thickness, however, less than 10% which is to within the experimental error of the accuracy, where the scaling exponents were also determined. Moreover, such a variation takes place over a range of film thicknesses sufficient to establish the asymptotic value  $\rho_{sf} \approx 0.7$ .

Nevertheless, discrepancies of the deposition details arising from precise control of the deposition rate during initial stages of growth, and frequent interruptions of the growth processes in order to prevent sample heating, could alter the mode of film growth by itself leading to the inconsistency  $z \neq H/\beta$  [13]. From the experimental relations  $\sigma = (0.385)h^{0.376}$ ,  $\xi = (5.5)h^{0.28}$ , and Eq. (4) for  $L \rightarrow +\infty$ , with  $H = 0.85$ , we calculated in Fig. 4 the local slope as a function of film thickness  $h$ . As can be observed, the local surface slope changes as an overall over the range 10–1000

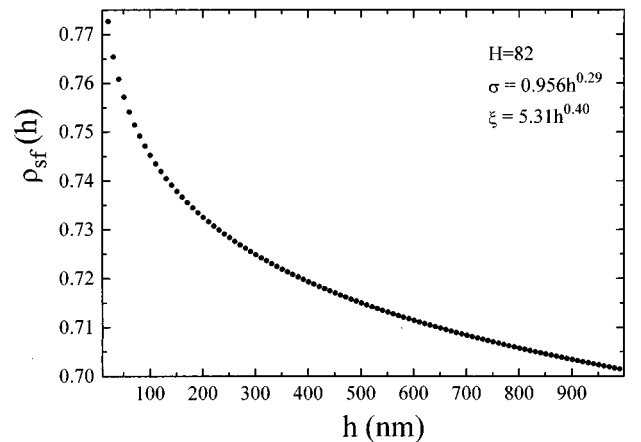


FIG. 3. Schematics of the rms local surface slope  $\rho_{sf}$  vs  $h$  (for  $L \rightarrow +\infty$ ) for the silver films, where  $z \approx \beta/H$  with  $a_0 = 0.3$  nm,  $\sigma = (0.956)h^{0.29}$ ,  $\xi = (5.31)h^{0.40}$ , and roughness exponent  $H = 0.82$ .

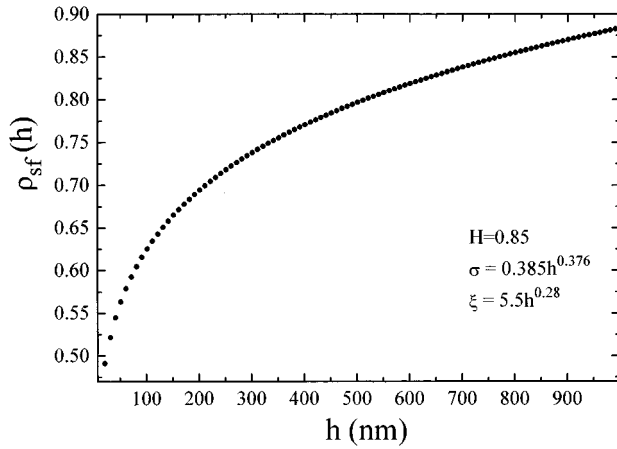


FIG. 4. Schematics of the rms local surface slope  $\rho_{sf}$  vs  $h$  (for  $L = +\infty$ ) for the silver films where  $z \neq \beta/H$  with  $a_0 = 0.3$  nm,  $\sigma = (0.385)h^{0.376}$ ,  $\xi = (5.5)h^{0.28}$ , and  $H = 0.85$ . The local slope increases following a power law behavior  $\propto h^{0.42}$ .

nm closely by more than 60% following a power law  $\rho \propto h^{0.42}$ , since  $(\beta z - H)/H \approx 0.42$ .

In the system under discussion, Schwoebel barriers (which usually exist at the step edge of well-defined terraces preventing step-down diffusion and resulting in violation of the self-affine hypothesis) are unlikely to be present due to the different observed morphologies [see scanning tunnel microscope (STM) images in Fig. 1 of Ref. [13]] than that of large scale pyramids characterizing growth with  $H = 1$  [19]. In addition, in our polycrystalline system, with randomly oriented grains, Schwoebel effects are rather unlikely to occur. However, if Schwoebel barriers are present, the slope of the pyramids remains constant for considerable barriers while it increases as a power law for weak barriers [20].

Equation (4) in the non-self-affine limit  $H = 1$  yields the logarithmic behavior  $\rho|_{H=1} = (\sigma/2^{1/2}a\xi)(\ln[1 + aQ_c^2\xi^2] - 2a)^{1/2}$  for  $L \gg \xi$ . On the other hand, at  $H = 1$  the self-affine correlation  $C(r) \sim e^{-(r/\xi)^{2H}}$  [15] yields the Gaussian roughness spectrum  $\langle |z(q)|^2 \rangle_g = [A/(2\pi)^6] \sigma^2 \xi^2 e^{-q^2 \xi^2/4\pi}$  with an associated rms local surface slope  $\rho_g = 2\pi^{1/2}(\sigma/\xi)[1 - e^{-Q_c^2 \xi^2/4\pi} - (Q_c^2 \xi^2/4\pi)e^{-Q_c^2 \xi^2/4\pi}]^{1/2}$ , at  $L \gg \xi$ . For  $\xi \gg a_0$ , we obtain in both cases  $\rho_g \approx 2\pi^{1/2}(\sigma/\xi)$  and  $\rho|_{H=1} \approx (\sigma/a\xi)\ln^{1/2}(\xi/a_0)$ . For  $\sigma \propto h^\beta$ ,  $\xi \propto h^{1/2}$ , and  $\beta = 1/z$  ( $H = 1$ ) [19], the Lorentzian model yields the logarithmic time dependence  $\rho|_{H=1} \approx \ln^{1/2}(t)$  which compares to the behavior observed in the anomalous scaling regime [9], while the Gaussian model yields  $\rho_g \sim \text{const}$ , which compares to the behavior observed in unstable growth with considerable Schwoebel barriers [20].

In conclusion, we investigated properties of the rms local surface slope of self-affine rough surfaces. Our calculations were based on phenomenological surface models which despite their *ad hoc* nature can capture the correct self-affine scaling behavior, and compare reasonably well in many cases with real data. Moreover, in terms of these models, analytically solvable expressions of the rms local slope were obtained which were also in agreement with the time invariance required in self-affine growth. Application of our results to the case of heteroepitaxially grown metal films enabled the estimation of the rms local slope over a wide range of film thicknesses (10–1000 nm). Finally, discrepancies in the deposition details can have great impact on the mode of film growth by itself, which is revealed through the inconsistency relation  $z \neq H/\beta$  among the scaling exponents or alternatively the thickness variance of the local surface slope.

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- [17] STM data in Ref. [13] for Ag films which are presented by squares and correspond to well controlled growth conditions. Independent measurement of the scaling exponents yielded  $H = 0.82 \pm 0.05$ ,  $\beta = 0.29 \pm 0.06$  ( $H/\beta = 2.8 \pm 0.6$ ), and  $z = 2.5 \pm 0.5$  in the thickness range 10–1000 nm.
- [18] STM data in Ref. [13] for Ag which are presented by circles and correspond to less controlled growth conditions. Silver films grown under these conditions revealed lower roughness amplitudes and correlation lengths, as well as the scaling exponents  $H = 0.85 \pm 0.09$ ,  $\beta = 0.376 \pm 0.007$ , and  $z = 3.6 \pm 0.3$ .

- ( $H/\beta = 2.3 \pm 0.25$ ) in the thickness range 10–1000 nm.
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